IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION OF: GROUP: 1793

Takuji IKEDA, et al.

SERIAL NO: 10/579,720   EXAMINER: Hoban, Matthew E.

FILED: May 17, 2006

FOR: HIGH SILICA CDS-1 ZEOLITE

DECLARATION UNDER 37 C.F.R. § 1.132

COMMISSIONER FOR PATENTS
ALEXANDRIA, VIRGINIA 22313

Sir:

Now comes Takuji Ikeda who deposes and states that:

1. I am a graduate of Tsukuba University and received my Ph.D degree in the year 1999.

2. I have been employed by National Institute of Advanced Industrial Science and Technology for 8 years as a researcher in the field of material science.

3. The following remarks are made in explanation and support of the patentability of the above-identified application:

The present Inventor respectfully submits that a crystalline layered compound (=PLS-1) defined in claim 1 of the present application and FLS (magadiite) of the prior art (=Reference A) are not the same.

With regard to FLS, as a reference other than Reference A, I have found a reference (=Reference B, Langmuir, Vol. 18, No. 10, 4103-4110, 2002), wherein FLS is described as FLS1.

There are some differences between PLS-1 and FLS/FLS1, and also CDS-1 and calcined FLS/FLS1 as follows.

1) Difference 1: Local structure by solid NMR

In the first place, with regard to the spectra of ^{29}Si MAS-NMR, the spectrum of PLS-1 of the present invention is different from that of FLS/FLS1 of References A/B.

In the second place, with regard to relative intensity ratio of Q^3 spectrum on $-94 \sim -107$ ppm and Q^4 spectrum on $-107 \sim -125$ ppm, Q^3/Q^4 ratio of PLS-1 is different from that of FLS/FLS1.

In the case of PLS-1 demonstrated in Fig. 4 of the present application, Q^3/Q^4 ratio of PLS-1 is higher compared with that of FLS/FLS1 (shown by a red arrow, in Fig.5,c in Reference A and Fig.5,c in the Reference B), which are evidences showing the amounts of Si-OH on the surface of crystals of these materials are different.

$Q^3/Q^4 = 0.714$ ($Q^4/Q^3 = 1.40$) of PLS-1 (See Fig. 1,[c] of Reference 1)

$Q^3/Q^4 = 0.26$ of FLS (See Reference A, line 9, left column, p.1617)

$Q^3/Q^4 = $ about 0.389 ($Q^4/Q^3 = 2.57$) of FLS1 (See Reference B, line 12 from the bottom, right column, p.4105)

As shown in Fig. 5,d, e in the Reference B, calcined FLS 1 shows <u>a apparent shoulder peak</u> in Q3 region (-94~-107 ppm)(shown by blue arrow) [→ this is very important] , which seems to be resulted from the incompleteness of dehydration and polycondensation and means that the skeleton of FLS1 has a structure, wherein (SiO$_4$) Si groups thereof are partially not closed, and <u>has many remained Si-OH groups</u>. This is essentially different from that of the ideal framework of zeolite.

In general, the material containing many $-$OH groups shows hydrophilic property which is specific to zeolite and the material containing less$-$OH groups such as CDS-1 shows hydrophobic property. Therefore, these materials are to be essentially different products.
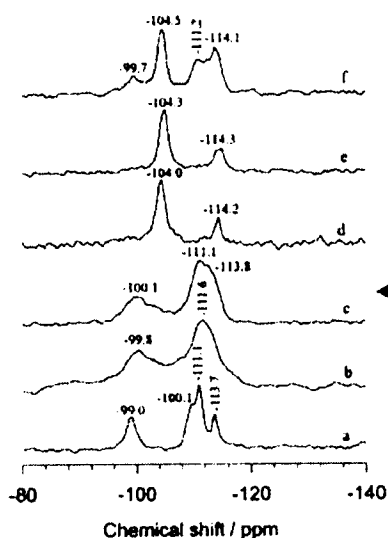
Fig.5 in Reference A   Fig.5 in Reference B

Fig. 5 ^{29}Si MAS NMR spectra of (a) Na-magadiite and (b–f) resulting phases obtained after reaction at 150°C for different times. See Fig. 2 for details.
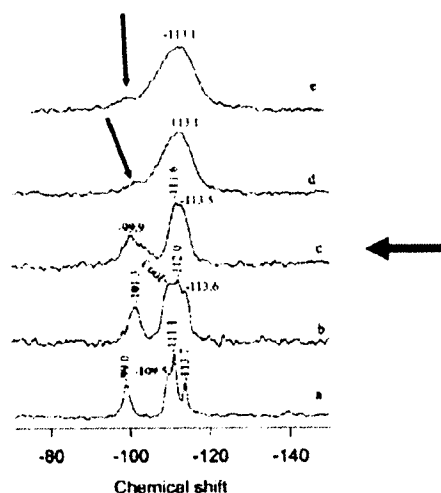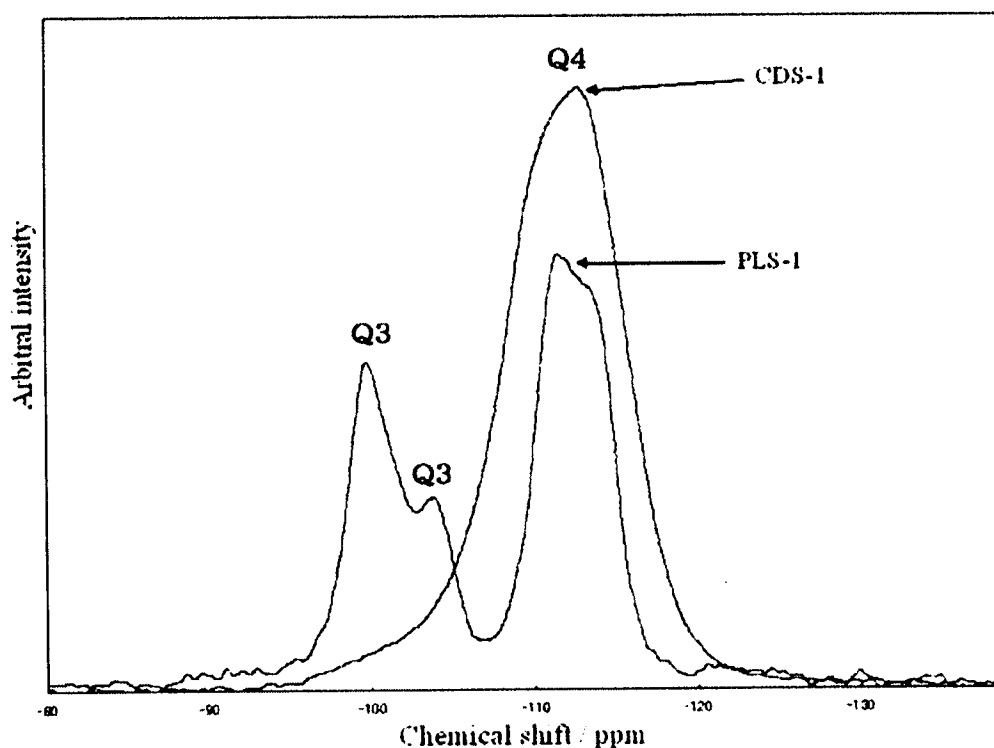
Figure 5. ^{29}Si MAS NMR of (a) Na-magadiite and (b) H-magadiite. Spectrum c corresponds to the FLS1 phase prepared at 150 °C for 5 days, then calcined at (d) 500 and (e) 700 °C.

^{29}Si MAS-NMR spectra of PLS-1 and CDS-1 (Fig.4 in this application)

2) Difference 2: Information of the micro pore by nitrogen adsorption method

In the Reference B, as a evidence for proving the calcined FLS-1 is microporous material, the average pore size is analyzed by nitrogen adsorption method, and it is revealed that the average pore size of calcined FLS-1 is 0.88 nm as shown in Fig. 10 in the Reference B. On the other hand, the average pore size of CDS-1 is 0.48 nm (the primary peak) and the secondary peak shows only 0.64 nm, as shown in Fig. 8 in the present application.
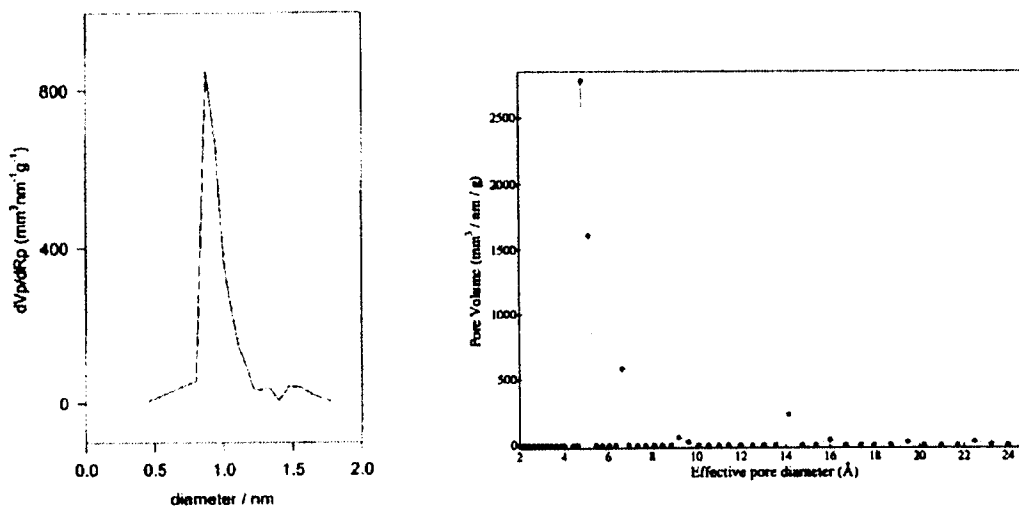
Figure 10. Pore size distribution of FLS1 calcined at 500 °C. using the MP method.

Fig.10 in Reference B   CDS-1 (Fig.8 in this application)

The crystal structure of CDS-1 is shown in Fig. 9 in the present application and is made clear in our publication (= T. Ikeda et al., Angew. Chem. Int. Ed., 2004, 43, 4892-4896: Reference 1).

Namely, the pore size of calcined FLS-1 is apparently larger than that of CDS-1.

The pore size of calcined FLS-1 is apparently larger than the effective pore size of CDS-1, which is 0.47nm as maximum pore size that can be calculated from the crystal structure shown by considering effective ion radius (1.35 Å) of oxygen described in Reference 2 (Atlas of Zeolite Framework Types, 6th revised edition, 2007, Ch. Baerlocher, W. M. Meier and D. H. Olson, ISBN: 978-0-444-53064-6).

The crystal structure of the CDS-1 is registered as the topology of CDO (CDS-One) by International Zeolite Association (IZA) (Reference 2, 3), which is in conforming to IUPAC, wherein the maximum pore size of CDS-1 is described as 0.47nm(Reference 2: Atlas of Zeolite Framework Types, 6th revised edition, 2007, Ch. Baerlocher, W. M. Meier and D. H. Olson, ISBN: 978-0-444-53064-6, Reference 3: http://izasc.ethz.ch/fmi/xsl/IZA-SC/ftc_tm.xsl?-db=Atlas_main&-lay=tm&STC=CDO&-find).

Further, with regard to the pore volume measured by nitrogen adsorption method, the pore volume of CDS-1 is calculated to be about 0.6 ml/g demonstrated from the present application and in the Reference 1, but the pore volume of calcined FLS1 is only 0.317 ml/g as maximum as demonstrated in Table 1 in Reference B.

With regard to zeolites obtained from same precursors, these zeolites should have almost similar average pore size and pore volume.

Table 1. Surface Areas of FLS1 Calcined at Different Temperatures Calculated Using the BET Equation[a]

sample	S_{BET} (m^2 g^{-1})	C_{BET}	corr coef	pore vol[c]
H-magadiite	40	137	0.999 98	
FLS1	90	128	0.999 85	0.141
FLS1(300)[b]	120	−316	0.999 46	0.114
FLS1(400)	306	−189	0.999 39	0.212
FLS1(500)	436	−180	0.999 68	0.317
FLS1(700)	307	−106	0.999 55	0.205
FLS1(800)	270	−134	0.999 58	0.181
FLS1(900)	95	282	0.999 88	0.115

[a] The corresponding CBET and correlation coefficients are also reported. [b] Values in parentheses indicate the calcination temperatures (°C). [c] mL (liquid nitrogen) g^{-1}.

Table 1 in Reference B

In view of the above, we can understand clearly that PLS-1 is different from FLS/FLS 1, and that CDS-1 is different from calcined FLS/FLS1.

XRD can show average structure of the materials, however, for example, it is impossible to know whether the covalent bonds of –Si–O–Si– are locally cleaved or not.

The calcined FLS/FLS1 is believed to be not a complete zeolite structure defined as CDO type topology as CDS-1 of the present invention, but to be a disordered structural material which is due to incompleteness of the dehydration and condensation between the layers of the precursor FLS1.

I hereby certify that CDS-1 obtained by calcination of precursor PLS-1 is different from calcined FLS1 which is synthesized by Kooli, and the precursor PLS-1 is different from FLS1.

4. The undersigned petitioner declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

5. Further deponent saith not.

Takuji Ikeda
Signature

2, July, 2010
Date

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